



Swelling of shale in supercritical carbon dioxide



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ABSTRACT

Shale gas is an unconventional but promising natural resource. One means of producing this gas is by injecting CO₂ into shale formations, and this technique has received widespread attention of late. This method not only allows the possibility of storing CO₂ in shale formations but also enhances the gas recovery process. The swelling of the shale matrix caused by CO₂ adsorption has important consequence with regard to the production of shale gas and the sequestration of CO₂ in shale formations. In this study, an apparatus was designed and used to measure CO₂-induced swelling in shale samples at temperatures between 308 and 348 K and pressures up to 15 MPa. The results show that CO₂-induced swelling occurs in shale samples. With increasing CO₂ pressure, the swelling of shale samples initially increases and then lessens. With increasing CO₂ temperature, the maximum swelling of the shale gradually decreases. The strain induced in the shale during this process in response to a constant CO₂ pressure can be divided into three regions: transient shrinkage, slow swelling and stable strain. The results of calculations employing a simplified local-density model were in agreement with the experimental data obtained from CO₂-induced swelling in shale. All experimental samples exhibited anisotropic strains in response to CO₂ injection, with the strains always being less in the direction parallel to the bedding plane.

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1. Introduction

Shale gas is an unconventional but promising gas resource that has been used with significant success in the United States of America (EIA, 2013). The extraction of this resource requires hydraulic fracturing and horizontal drilling in shale formations together with the use of large amounts of water. Owing to the high clay content of the shale gas reservoirs in China (Wang et al., 2012), and it is almost certain that drilling will be necessary. One associated issue is the need to control the swelling caused by water injected into clay-rich shale. Many researchers have instead focused on using CO₂ injection for fracturing (Tsuyoshi et al., 2012). This process could, in fact, represent a new method of carbon capture, utilization and storage, such that CO₂ is sequestered in shale formations while enhancing shale gas recovery (Liu et al., 2013). The CO₂-induced swelling of shale matrices has important implications in selecting the methods used for the production of

shale gas and the storage of CO₂ in shale formations.

Over the past decade, several studies have focused on swelling induced by the adsorption of gases. Day et al. (2007) measured the swelling caused by the introduction of CO₂ into Australian coals and showed that the maximum volumetric strain was between about 1.7 and 1.9%. Recently, Ferian et al. (2014) studied the strain induced by CO₂ in low rank coals and reported that the maximum volumetric strain was on the order of 1.65%. Other researchers have proposed a theoretical model to describe CO₂-induced swelling in coal (Pan and Luke, 2007; Pan & Connell, 2012; Pongtorn et al., 2014). The above works focused on CO₂-induced swelling in coal but the swelling of shale during adsorption CO₂ should also be addressed. However, to our best knowledge, there have been very few studies on the swelling of shale by CO₂.

The temperatures of high organic content shale formations range from 370 to 550 K, while the pore pressure in such formations is in the range of 15–20 MPa. Carbon dioxide will exist as a supercritical state under these conditions, given that the critical pressure of carbon dioxide is 7.38 MPa and the critical temperature is 304.13 K. The supercritical state is a phase that a substance will transition to at temperatures and pressures beyond the critical pressure. This state has unique characteristics, including low

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viscosity and high diffusivity. [Chalmers and Bustin \(2007\)](#) reported that the CO₂ storage capacity of shale reservoirs based on the injection of CO₂ was linked to the physical sorption of CO₂ in the shale. Theoretical research has shown that the surface potential energy of the shale is reduced by the adsorption of CO₂. If the specific surface energy is decreased in this manner, the shale should swell to increase its surface area ([Scherer, 1986](#); [Pan and Luke, 2007](#)). This swelling could have serious consequences for CO₂ sequestration or hydraulic fracturing using CO₂ or water, and will change the formations. Among the possible consequences are closing of cracks (and thus reduced porosity) and reduced permeability of the shale formation. Therefore, it is important to carry out research on CO₂-induced swelling of shale. [Busch et al. \(2008\)](#) found that CO₂ might affect both the porosity and permeability of shale. [Kumar et al. \(2010\)](#) used CH₄, He and CO₂ to measure the permeability of shale, and indirectly showed that the adsorption of CO₂ will generate strain. Recently, [Richard et al. \(2011\)](#) investigated the pore structure changes caused by CO₂ injection into shale, and found that carbonate minerals enter into solution and that pore structure changes are not observed at high CO₂ pressures. However, data were not acquired simultaneous with the injection process during these experiments.

In the present work, an apparatus with the ability to measure the swelling in shale induced by CO₂ was designed and built. This device was used to measure the adsorption capacity and CO₂-induced swelling of shale samples at temperatures between 308 and 348 K and pressures up to 15 MPa. Both the CO₂-induced swelling and adsorption capacity were assessed based on the resulting experimental data and the resulting strain and strain anisotropy values were studied.

2. Theories

2.1. CO₂-induced swelling description

Two methods were employed to describe CO₂-induced swelling in our samples. In the first, a theoretical model was used to describe the adsorption-induced volumetric strain in samples ([Wu et al., 2005](#); [Zhou et al., 2011](#); [Pan and Luke, 2007](#)). This method involved numerous parameters. The second approach used the adsorption form of an equation describing the volumetric changes in sample *s* ([Levine, 1996](#)).

In the following discussion, we use the second approach to describe the volumetric changes in shale samples. It is known that gases will be present in a shale formation in the 370–550 K temperature range ([Thomas et al., 2013](#)), although CO₂ will be in the supercritical state. Thus, the simplified local-density (SLD) model ([Pongtorn et al., 2012](#)) can be used to describe CO₂ adsorption on shale.

In the case of an equilibrium adsorption state, the chemical potential of the adsorbed phase is equal to the chemical potential of the bulk phase. The chemical potential of the adsorbed phase can be expressed as the sum of the fluid–solid and fluid–fluid potentials as follows ([Polanyi, 1963](#)):

$$\mu_{bulk} = \mu_{ff}(Z) + \mu_{fs}(Z) \quad (1)$$

where the subscript “bulk” indicates the bulk fluid, the subscript “fs” indicates the fluid–solid interaction, the subscript “ff” indicates the fluid–fluid interaction and *Z* is the vertical distance between the fluid molecules and the solid surface.

The fugacity value can be used to calculate μ_{bulk} as follows ([Polanyi, 1963](#)):

$$\mu_{bulk} = \mu_0(T) + RT \ln(f_{bulk}/f_0) \quad (2)$$

where the subscript “0” indicates an arbitrary reference state. Analogously, $\mu_{ff}(Z)$ can be written as ([Polanyi, 1963](#)):

$$\mu_{ff}(Z) = \mu_0(T) + RT \ln(f_{ff}(Z)/f_0) \quad (3)$$

where $f_{ff}(Z)$ is fluid fugacity at position *Z*.

The fluid–solid potential is given by [Polanyi \(1963\)](#):

$$\mu_{fs}(Z) = A_V \psi(Z) \quad (4)$$

where A_V is Avogadro's constant and $\psi(Z)$ is the fluid–solid interaction of a fluid molecule.

Substituting Eqs. (2)–(4) into Eq. (1) provides the balance for adsorption:

$$f_{ff}(Z) = f_{bulk} \exp(-\psi(Z)/kT) \quad (5)$$

f_{bulk} is calculated by equation of state (EOS). The selection of EOS is the key factor of SLD model. The Elliott-Suresh-Donohue (ESD) EOS is used in this study. The fugacity of bulk fluid is used in ESD-EOS ([Elliott et al., 1990](#)):

$$\ln f = -\frac{4}{19} c \ln(1 - 1.9\eta) + \frac{4c\eta}{(1 - 1.9\eta)} - \frac{9.5q}{1.7745} \ln(1 + 1.7745Y\eta) - \frac{9.5qY\eta}{(1 + 1.7745Y\eta)} - \ln \frac{V}{RT} \quad (6)$$

where $V = 1/\rho$ is the molar volume.

The term $\varphi(z)$ is related solely to the distance, *Z*, and the fluid–solid interaction between the fluid molecules and the solid surface may be written as ([Duong, 1998](#)):

$$\varphi'(z) = 2\pi\rho_c \varepsilon_{sf} \sigma_{sf}^2 \Delta \left(\frac{2}{5} \left(\frac{\sigma_{sf}}{z} \right)^{10} - \left(\frac{\sigma_{sf}}{z} \right)^4 - \left(\frac{\sigma_{sf}^4}{3(z + 0.61\Delta)^3} \right) \right) \quad (7)$$

where $\rho_c = 114nm^{-3}$ is the average density of the atoms in the sample, $\Delta = 0.335nm$ is the distance between adjacent sample layers, ε_{sf} is the fluid–solid interaction energy and $\sigma_{sf} = 0.356nm$ is the distance between the adsorption phase and the carbon interplanar.

In this model, the adsorption potential energy of the adjacent wall surfaces in the micropores is superimposed. In addition, the fluid–solid interactions in the model are accounted for through a potential energy function ([Subramanian et al., 1995](#)):

$$\varphi(z) = \varphi'(z) + \varphi'(H - z) \quad (8)$$

where *z* is the vertical distance between gas molecules and one wall surface and the value of $\varphi(z)$ can be obtained by inserting Eq. (8) into Eq. (7). The value of $f_{ff}(Z)$ can be obtained by inserting the values for $\varphi(z)$ and f_{bulk} into Eq. (5).

With regard to the adsorption fluid, the fugacity for the fluid–fluid interaction $f_{ff}(Z)$, is given by the following equation ([Wang and Johnson, 1999](#)).

$$\ln f_{ff}(Z) = -\frac{4}{19} c \ln(1 - 1.9\eta) + \frac{4c\eta}{(1 - 1.9\eta)} - \frac{9.5q}{1.7745} \ln(1 + 1.7745Y(z)\eta) - \frac{9.5qY(z)\eta}{(1 + 1.7745Y(z)\eta)} - \ln \frac{V}{RT} \quad (9)$$

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